

Weather Extremes Led to Large Variability in O₃ Pollution and Associated Premature Deaths in East of China

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As global warming intensifies, hot extremes and heavy precipitation frequently happen in East of China. Meanwhile, severe surface ozone (O_3) pollution resulting from the interactions of anthropogenic emissions and meteorological conditions also occur more frequently. In this study, we quantified the impact of weather extremes on ground-level O3 concentration during the summers of 2015-2021 and associated premature deaths in East of China. The O3 pollution influenced by hot extremes [maximum 8-h average O_3 concentration (MDA8 O_3) = 152.7 µg m⁻³] was 64.2% more severe than that associated with heavy rain (MDA8 $O_3 = 93 \mu g m^{-3}$) on the daily time scale. The compound hot and dry air extremes had a larger impact, and the associated MDA8 O₃ could be up to 165.5 μ g m⁻³. Thus, weather extremes could drastically perturb the O₃ level in the air to exhibit large variability. Based on GEOS-Chem simulations with fixed anthropogenic emissions, forcing of weather extremes could successfully reproduce the large daily variability of O3 concentration because the weather extremes significantly influenced the physicochemical processes in the atmosphere. Furthermore, hot extremes magnified the single-day O₃-related premature death to 153% of that under other-condition events, while heavy rain events decreased it to 70% in East of China. The findings of the present study have the potential to promote daily to weekly O_3 forecasts and further improve our comprehensive understanding of the health effects of weather extremes and air pollution.

Keywords: weather extreme, high temperature, heavy rain, O₃ pollution, premature mortality

INTRODUCTION

Weather extremes have been occurring more frequently and are posing huge threats to human society, infrastructure, and the natural environment (Zhang Q. et al., 2017; Chen and Sun, 2021; Wang et al., 2022). The sequential flood-heatwave disasters in Japan during 2018 caused thousands of deaths in a single week (Kim et al., 2019). In 2021, an extreme precipitation event hit the Henan Province in China and resulted in more than 300 deaths and an economic loss of 17.7 billion dollars (World Meteorological Organization, 2021). In addition, the heatwave in the summer of 2003 led to

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1

about 70,000 deaths in Europe (Robine et al., 2008). Furthermore, largest warming (particularly increases in maximum air temperature) and enhanced heavy precipitation were projected to occur in East of China in most of the coming decades (Zhou et al., 2014; Li et al., 2021; Qiao et al., 2021; Yang et al., 2021).

In recent years, ground-level ozone (O₃) has exceeded fine particulate matters to become a primary air pollutant that affects East of China (Wang et al., 2017; Fu et al., 2019). Meanwhile, the O₃ pollution in the summer was the most serious and would have caused a greater impact on human health. Because anthropogenic emissions remain approximately invariant on the daily time scale, the daily variability of O3 concentration was mainly dominated by weather conditions (Han et al., 2020; Yin and Ma, 2020; Tang G. et al., 2021). Large concentrations of surface O₃ were observed to occur frequently with high air temperature, strong solar radiation, and low air humidity (Pu et al., 2017; Li et al., 2022). The relationships between high ozone days and weather extremes in the eastern U.S. have been analyzed, and Zhang H. et al. (2017) found that high ozone extremes were most sensitive to daily maximum air temperature and minimum relative humidity. Based on simulations of the Weather Research and Forecasting model coupled with Chemistry (WRF-Chem), the O3 concentration in the U.S. associated with heat waves and atmospheric stagnation proved to be much larger than that under non-extreme weather conditions in the present and also in the future. Moreover, compound weather events had a larger impact than a single event because the temperature was noticeably higher and the wind speed was abnormally weaker (Zhang et al., 2018). However, quantitative studies on the impacts of weather extremes on O₃ pollution in East of China are far less than sufficient to the best of our knowledge.

Short-term exposure to peak levels of ozone can largely affect the lungs, the respiratory tract, and the eyes, and increase susceptibility to inhaled allergens. In the worst situation, it can even lead to premature mortalities (Chen et al., 2017; Orellano et al., 2020). Even when O3 concentration was lower than the level influencing pulmonary function, the blood pressure of healthy adults could still increase and possibly affect their cardiovascular health (Day et al., 2017). Fenech et al. (2019) examined the meteorological drivers associated with two five-day episodes of air pollution in 2006 and found about 70 daily premature deaths because of short-term O₃ exposure across the UK. In China, the number of premature deaths attributed to shortterm O₃ exposure increased by 9,100 from 2013 to 2018 in 74 key cities, and the national premature mortalities in 2018 were 6.32 \times 10⁴ (Wang et al., 2021). With every $10 \,\mu\text{g/m}^3$ increase in ozone, the rates of respiratory deaths and non-accidental deaths respectively increased by 2.22 and 0.05% in highly O₃-polluted areas of China (Lei et al., 2019). As shown clearly from site observations, large daily variabilities of O₃ concentration existed (Supplementary Figure S1), and must have resulted in evident day-to-day differences in premature mortalities.

At present, few studies have been conducted to quantify to what extent weather extremes (e.g., hot, hot-dry, and heavy rain events) influence O_3 concentration and their associated health effects in East of China. Accurate quantification of large O_3 variability and premature deaths associated with different weather extremes could not only provide scientific support for O_3 pollution forecasts but also contribute to a comprehensive understanding of the health effects of weather extremes and air pollution.

DATA AND METHODS

Observations

The daily meteorological data with a horizontal resolution of 0.5° latitude by 0.625° longitude from 1981 to 2021 were derived from the Modern-Era Retrospective analysis for Research and Applications version 2 (MERRA2) dataset (Gelaro et al., 2017). The data included daily maximum temperature (Tmax), surface relative humidity, precipitation, planetary boundary layer (PBL) height, and downward solar radiation. In addition, the Tmax and precipitation data from the gridded daily observation dataset over China region (CN05.1) were downloaded to verify the impact of weather extremes on O₃. Surface O₃ concentrations have been observed and widely implemented since 2015 in China. The impact of weather extremes on O₃ during 2015–2021 were studied. Ground-level O3 concentrations from 2015-2021 were downloaded from https://quotsoft.net/air/. The daily maximum 8-h average O₃ concentration (MDA8 O₃) was used to represent ozone pollution condition.

Weather Extremes

Following Lu et al. (2016) and Zhang et al. (2011), we used percentile-based indices to describe three types of weather extremes as follows:

- Warm temperature events indicated weather conditions with Tmax higher than the 90th percentile threshold during 1981–2010 (hereafter called TX90).
- (2) Compound hot and dry air events indicated weather conditions with Tmax higher than the 90th percentile threshold and relative humidity smaller than 10th percentile during 1981–2010 (hereafter called TX90H10).
- (3) Heavy precipitation events indicated weather conditions with daily precipitation higher than the 95th percentile threshold during 1981–2010 (hereafter called R95).

All other weather conditions, except for the aforementioned three weather extremes in the summer, were defined as other-condition events (hereafter called OCE).

GEOS-Chem Simulation

A global 3-D chemical transport model (GEOS-Chem) was implemented to simulate surface ozone concentration in East of China. The GEOS-Chem model includes fully coupled O_3 -NOx-hydrocarbon and aerosol chemistry with more than 80 species and 300 reactions (Bey et al., 2001). The major physical-chemical processes used for the budget diagnostics included chemical reaction, transport, PBL mixing, convection, and their summary within the PBL (Liao et al., 2006).



boxes represent the location of North China, Yangtze River Delta, and Pearl River Delta.

The GEOS-Chem model could well reproduce the dailyinterannual variation and spatial distribution of surface ozone concentration (Yin and Ma, 2020; Ma and Yin, 2021). In this study, we also evaluated the performance of the GEOS-Chem model. Observations and model results during the summer of 2020 and extreme weather events during 2015-2021 were shown in Supplementary Figure S1. Statistical metrics such as correlation coefficient (Cor), normalized mean bias (NMB), and normal mean error (NME) were calculated. The spatial correlation coefficient between observations and the model simulation, respectively, is 0.87 and 0.82 during the summer of 2020 and extreme weather events, indicating that the simulated MDA8 O3 agreed well with the observed distribution. Moreover, NMB was 0.49% (4.9%) and NME was 10.2% (11.2%) during the summer of 2020 (extreme weather events), indicating a good capability of the GEOS-Chem model for the simulation of MDA8 O₃ concentration both on a daily time scale and weather extremes. However, the GEOS-Chem model was short of reproduction when the MDA8 $O_3 > 150 \,\mu g \,m^{-3}$, i.e., underestimating the severest surface O₃ caused by weather extremes (Supplementary Figure S1b).

Premature Mortality

The Environmental Benefits Mapping and Analysis Program-Community Edition (BenMAP-CE) has been used to calculate the premature mortality due to short-term exposure to ozone pollution (Fenech et al., 2019). The formulas are as follows:

$$M = Y_0 P\left[\frac{(R-1)}{R}\right],\tag{1}$$

$$\mathsf{R} = \exp^{\beta \left(\mathsf{C} - \mathsf{C}_f\right)},\tag{2}$$

where *M* is the all-cause premature mortality attributed to ambient ozone, Y_0 is the baseline mortality rate due to a specific disease category, and *P* is the population. The value of Y_0 from 2015 to 2019 could be obtained from the China Public Health and Family Planning Statistical Yearbook, and Y_0 for 2020–2021 was set to be the same as that in 2019. The gridded population data were downloaded from the Socioeconomic Data and Applications Center. *R* is the relative risk for a specific disease, which can be calculated by utilizing the integrated exposure response (IER) model. β is the estimated slope of the log-linear relationship between ozone concentration and allcause mortality. Ye et al. (2020) conducted a meta-analysis of



the associations between short-term ozone exposure and human mortality in China. Their result ($\beta = 0.0003992$) was used in the present study. *C* is the observed ozone concentration (unit: $\mu g m^{-3}$), and C_f is the theoretical minimum concentration is set to 70 $\mu g m^{-3}$ (Wang et al., 2021).

LARGE O₃ VARIABILITIES DUE TO WEATHER EXTREMES

During the summers of 2015–2021, the occurrence frequencies of hot extreme and heavy rain (i.e., TX90 and R95) were higher than those during 1981-2014 in most of the stations in East of China (Figures 1A,C). There were 10.2 TX90 days per summer in East of China (8.3 in NC, 8.4 in YRD, and 15 days in PRD) during 2015-2021, while the number of R95 days was 5.1 per summer (5, 6.1, and 4 days in NC, YRD, and PRD, respectively). The mean of Tmax values accompanied with the occurrences of TX90 and R95 events were 37.3 and 28.9°C, respectively. The intensities of precipitation during TX90 and R95 were 0.4 and 34 mm/d, respectively. This result indicated significant differences in meteorological conditions (above 95% confidence level). Furthermore, the mean PBL heights on the days of TX90 and R95 were 585.7 m (12.7% higher than OCE) and 442.6 m (14.8% lower than OCE) and the surface incoming shortwave fluxes were 9.7×10^5 J m⁻² (26% stronger than OCE) and 4.8×10^5 J m⁻² (37.7% weaker than OCE), respectively (Supplementary Figure S2). As a compound extreme weather, TX90H10 represented the

meteorological conditions with both hot temperature (mean Tmax = 37.5° C) and dry air (mean relative humidity = 29.4%). There were 2.8 TX90H10 days per summer in East of China (2.1, 2.2, and 7 days in NC, YRD, and PRD, respectively, as shown in Figure 1B). Hot temperature contributed to efficient photochemical reactions and natural precursor emissions (Pu et al., 2017). Dry air and strong solar radiation also helped to enhance photochemical reactions in the air (Zhang and Wang, 2016; Lu et al., 2019). Detected by tethered balloons, higher PBL height with stronger free convection condition were found to be beneficial for the aggravation of ozone pollution in China (Tang R. et al., 2021). The impacts of precipitation on surface O_3 were relatively complex. In addition to temperature reduction and cloud shading accompanied with precipitation process, heavy rain itself could remove the stock of O_3 in the air (Meleux et al., 2007). Therefore, large daily differences in weather conditions potentially resulted in large variabilities of surface O₃ concentration.

To quantify the impacts of weather extremes, O₃ concentrations under TX90, TX90H10, and OCE during the summers of 2015-2021 were composited. The values were 152.7 μ g m⁻³, 165.5 μ g m⁻³, and 115.6 μ g m⁻³, respectively, in East of China (Figure 2). That is, the hot extremes could increase O3 concentration by 32.1% in East of China with respect to O₃ concentration under OCE conditions. When dry air was coexistent with hot temperature (i.e., compound extreme TX90H10), the anomalous percentage of O₃ concentration was raised to 43.2%. However, the mean O₃ concentration in R95 days was only 93 μ g m⁻³, indicating that the meteorological condition of heavy rain could reduce O₃ concentration by 19.6% (Figure 2). In general, the O₃-polluted condition corresponding to TX90 (TX90H10) was 51.7% (62.8%) worse than that corresponding to R95. In other words, weather extremes resulted in quite large variabilities of ground-level O3 concentration. In addition to the aforementioned observational analysis, the GEOS-Chem model was also driven by weather extremes but with fixed anthropogenic emissions to simulate O₃ concentration. Many previous studies pointed out that numerical models had limited capability for the simulation of weather extremes and air pollution in response to the extremes (Zhang and Wang, 2016; Zhang et al., 2018; Zhang et al., 2020).

Underestimation of high concentrations of O₃, as shown in Supplementary Figure S1B, is one of the possible reasons that the GEOS-Chem model failed to simulate the large percentage change of MDA8 O3 induced by weather extremes in East of China. Results of the present study indicate that, although the quantitative values simulated by the GEOS-Chem model were smaller, the significant influences of TX90, TX90H10, and R95 on O₃ concentration were well verified (Figure 2; Supplementary Figure S3). Similar analysis was also conducted for each individual year. The abnormal percentages of O₃ concentration remain steadily at around 30% under TX90, 40% under TX90H10, and -20% under R95 during the years of 2015-2021 (Supplementary Figure S3). These quasi-steady percentage changes would be helpful in forecasting O₃ pollution on daily to weekly time scales.



To explore the spatial heterogeneity of O₃ concentration, the impact of weather extremes in three severely O₃-polluted areas were analyzed. The summer-mean O₃ pollution was the severest in NC (145.7 μ g m⁻³), followed by that in YRD (115.9 μ g m⁻³) and PRD (91.2 μ g m⁻³). O₃ concentrations in all the three areas significantly exceeded the peak-season threshold of O3 concentration (i.e., $60 \,\mu g \,m^{-3}$) recommended by the World Health Organization (Zhu et al., 2022). Closely related to the regional O₃-polluted level, the O₃ concentrations under TX90 (TX90H10) were 191 (193.3), 154.6 (173.9), and 133.3 (143.3) μ g m⁻³ in NC, YRD, and PRD during the summers of 2015–2021 (Figure 2). Under the meteorological condition of heavy rain, however, the O3 concentrations were only 112.9, 84.3, and 76.0 μ g m⁻³ in NC, YRD, and PRD, respectively (Figure 2). Unlike absolute O₃ concentration, the anomalous percentage of O₃ concentration under TX90 compared to that under OCE was about 34% both in NC and YRD, but it increased to 60% in PRD (Supplementary Figure S4). This result indicated that hot extremes could induce a larger percentage increase of O₃ concentration compared to that in normal conditions in PRD than in the other two regions. The O₃ anomalous percentage under R95 was -20.9, -25.8, and -6.1% in NC, YRD, and PRD, respectively, during the summers of 2015-2021. These regional differences were also evident in the GEOS-Chem simulations (Supplementary Figure S4), and they probably had close relationships with local climate conditions and anthropogenic emissions.

Associated physical-chemical processes in East of China were analyzed based on the GEOS-Chem simulations (Figure 3). During OCE days, the mass fluxes of chemistry, transport, convection, and



PBL mixing were 6.36, -1.14, 0.44, and -2.14 Tons d⁻¹. In regard to chemical processes, high temperature could induce larger natural emissions of BVOCs and soil NOx, accelerate photochemical reaction (Lu et al., 2019), and enhance thermal decomposition of peroxyacetyl nitrate (PAN) to provide additional NO_x (Doherty et al., 2013). Conversely, heavy rain with more cloud cover could reduce the photolysis rate of NO2 (Lien and Hung, 2021). Meanwhile, the reduced solar radiation led to decreases in ozone production rate and natural emission (Jiang et al., 2021). Consequently, hot extremes led to almost double mass flux of chemistry $(11.32 \text{ Tons d}^{-1})$. In contrast, the chemical flux was largely suppressed by heavy rainfall in East of China (1.55 Tons d^{-1} , Figure 3). The fluxes of transport and mixing processes heavily depended on wind anomalies, free convection conditions, ozone level and its concentration gradient (Jiang et al., 2021; Tang G. et al., 2021). During TX90 days, the atmospheric circulation could transport more O3 pollutants under TX90 condition $(-3.07 \text{ Tons d}^{-1})$. However, the mass flux of transport turned out to be positive $(1.37 \text{ Tons d}^{-1})$ during heavy rainfall events, indicating that more O₃ pollutants remained in the air (Figure 3). Compared to that in the OCE days, the anomaly of PBL mixing flux was negative $(-0.69 \text{ Tons d}^{-1})$ due to hot temperature, while that under the influence of heavy rain was positive $(0.68 \text{ Tons d}^{-1})$. As shown in Figure 3, the quantity value of convection and its response to weather extremes were both tiny. In summary, when the weather was hot (hot-dry), 64.6% (72.5%) more O_3 pollutants were produced. However, the O_3 production was reduced by half during heavy precipitation events (Figure 3).

IMPACTS ON PREMATURE DEATHS

Weather extremes not only affected human health by basic meteorological factors (i.e., hot environment and rainfall intensity), but also modulated premature deaths associated with surface O₃ pollution. Short-term O₃ exposure could increase human premature deaths by elevating the risks of respiratory and cardiovascular diseases (Dong et al., 2016; Lei et al., 2019). To comprehensively understand the causes and consequences related to large variabilities of ozone pollution, the all-cause premature mortalities due to short-term O3 exposure were calculated using the BenMAP-CE approach. The number of accumulated premature mortalities in the summers of 2015–2021 was 13.8×10^4 (90% CI: $12.4-15.1 \times 10^4$) in East of China (Figure 4). Furthermore, the premature mortalities were greatly different between NC (5.6×10^4 , 90% CI: $5.0-6.1 \times 10^4$), YRD $(2.7 \times 10^4, 90\% \text{ CI: } 2.4-3.0 \times 10^4)$, and PRD $(0.3 \times 10^4, 90\% \text{ CI: } 2.4-3.0 \times 10^4)$ CI: $0.29-0.35 \times 10^4$). These obvious regional differences were mainly a result of local levels of O3 pollution and resident population. For example, C_f was set to 70 µg m⁻³, which did not frequently occur in PRD, and the premature mortalities were obviously lesser than in the other two regions (Supplementary Figure S5).

In addition to the spatial disparity, the aforementioned premature mortalities also varied greatly on a daily time scale. Hot temperature enlarged the daily death toll owing to short-term O₃ exposure to 153% of that under OCE conditions. Dry air would pile up another 23% increase in danger. However, heavy rain would decrease the O₃ concentration and thus cause fewer daily premature deaths, which was 70% under the OCE conditions. As shown in Figure 4, the premature mortalities due to O₃ exposure associated with TX90 (excluding TX90H10), TX90H10, and R95 in East of China were 1.56×10^4 (90% CI: $1.40-1.71 \times 10^4$), 0.77×10^4 (90% CI: 0.69-0.84 × 10⁴), and 0.43×10^4 10^4 (90% CI: 0.38–0.47 × 10⁴) persons, respectively. Although the impacts of weather extremes on short-term O3 exposure were significant, 79.7% of premature mortalities occurred without weather extremes because of a much larger OCE frequency (Figure 4). It is noted that while TX90 and R95 were not independent, there was only 1 day in seven summers in East of China when these two weather extremes happened simultaneously, which had little impact on total premature mortalities. The percentages were 82.3, 82.1, and 11.9% in NC, YRD, and PRD (Supplementary Figure S5), respectively. The small percentage in PRD was closely related to the relatively good air quality there, and indicated most remarkable impacts of weather extremes on premature mortalities due to short-term O₃ exposure.

CONCLUSION AND DISCUSSION

In this study, we quantified the impacts of weather extremes on surface O_3 pollution in East of China and their modulation of premature deaths caused by short-term O_3 exposure. Hot extremes (i.e., TX90 with Tmax = 37.3°C) could greatly enhance the mass fluxes of chemistry, which was favorable for

production of more O_3 pollutants (Figure 3). Although more O_3 could be removed by transport and PBL mixing processes, the summary mass fluxes were still 64.6% higher than those under other-condition events (i.e., OCE). Conversely, heavy precipitation (i.e., R95 with rain intensity = 34 mm/d) and associated meteorological conditions could obviously reduce the production of O_3 pollutants (-48.7% lower than that in OCE days). Consequently, the MDA8 O3 were 152.7 and $93 \,\mu g \,m^{-3}$ in East of China under TX90 and R95 conditions, respectively, which were 32.1% higher and 19.6% lower than their mean values in OCE days. The compound hot-dry extreme could result in a larger O₃ concentration that is 43.2% higher than that under the OCE conditions (Figure 2). Furthermore, the impacts of weather extremes on O₃ variability were geographically inhomogeneous. That is, a hot environment could lead to a larger deviation of O₃ concentration in PRD than in the other two regions, while the influence of heavy rainfall on O₃ concentration was the weakest in PRD. In addition to observational analysis, GEOS-Chem simulations were also conducted to verify the large variability in O₃ pollution driven by weather extremes. To enhance the reliability of the conclusion, the CN05.1 dataset was also employed to explore the impact of weather extremes on O₃ concentration. As shown in Supplementary Figure S6, identical results were obtained.

The health benefits of short-term O₃ exposure were also evaluated under different weather conditions. It is found that hot extremes increased the daily death toll to 153% of that under other-condition events. In contrast, the daily death toll decreased to 70% of that under other-condition events by heavy precipitation. The premature mortalities due to short-term O₃ exposure respectively associated with TX90 (excluding TX90H10), TX90H10, and R95 were 1.56×10^4 (90% CI: $1.40-1.71 \times 10^4$), 0.77×10^4 (90% CI: $0.69-0.84 \times 10^4$), and 0.43×10^4 (90% CI: 0.38–0.47 × 10⁴) persons in East of China (Figure 4). However, these results about premature mortality were preliminarily estimated by an idealized tool named BenMAP-CE, and they did not exactly correspond to the actual mortalities. It is imperative to check the relationships based on clinical cases and analyze the associated mechanisms. Furthermore, the weather extremes could also affect human health and lead to premature mortality by the meteorological conditions without O₃ pollution. Therefore, it is emergent to distinguish the health effects between weather extremes themselves and the associated O₃ extremes. More importantly, the new findings of the present study provides a more comprehensive understanding of health effects of weather extremes and air pollution.

Nowadays, daily to weekly forecasts of weather and air quality to a great extent depends on numerical models. It is widely known that weather models still have limited capability for forecasting weather extremes; that is, obvious biases exist in the numerical prediction (Huang and Ding, 2021). In this study, we find that the atmospheric chemical model failed to simulate the large percentage change of MDA8 O_3 induced by weather extremes in East of China, which implies the

imperative necessity to improve model performance for the simulation of extreme events by both numerical weather models and atmospheric chemical models. In addition, the mechanisms of how a specific meteorological element influenced the atmospheric physical-chemical processes (particularly fluxes of transport and PBL mixing) were still unclear and needed further studies. The observed quasisteady changes of MDA8 O_3 concentration influenced by weather extremes had the potential to promote daily to weekly forecasts and further support short-term control measures, such as limitations on industrial and traffic emissions.

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/**Supplementary Material**; further inquiries can be directed to the corresponding author.

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AUTHOR CONTRIBUTIONS

HW and BZ designed and supported this research. ZY and YW performed statistical and numerical studies. QH participated in analysis of health benefits. ZY prepared the manuscript with contributions from all co-authors.

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SUPPLEMENTARY MATERIAL

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